μSR Studies of Magnetic-Field-Induced Effects in High-T_c Superconductors

A. T. Savici,[1] A. Fukaya,[1] I. M. Gat-Malureanu,[1] T. Ito,[1] P. L. Russo,[1] Y. J. Uemura,[1,*] C. R. Wiebe,[1,2] P. P. Kyriakou,[2] G. J. MacDougall,[2] M. T. Rovers,[2] G. M. Luke,[2] K. M. Kojima,[3] M. Goto,[3] S. Uchida,[3] R. Kadono,[4] K. Yamada,[5] S. Tajima,[6] T. Masui,[6] H. Eisaki,[7] N. Kaneko,[7] M. Greven,[7] and G. D. Gu[8]

[1]*Dept. of Physics, Columbia University, New York, New York 10027*
[2]*Dept. of Physics and Astronomy, McMaster Univ., Hamilton, ON, Canada*
[3]*Dept. Physics, University of Tokyo, Tokyo 113-8656, Japan*
[4]*Institute of Materials Structure Science, KEK, Tsukuba, Japan*
[5]*Institute for Chemical Research, Kyoto University, Uji, Kyoto 611-0011, Japan*
[6]*ISTEC, Shinonome 1-10-13, Koto-Ku, Tokyo 135-0062, Japan*
[7]*Dept. of Applied Physics, Stanford University, Stanford, California 94305*
[8]*Brookhaven National Lab., Upton, New York 11973*

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Muon spin relaxation (μSR) measurements in high transverse magnetic fields ($\parallel \hat{c}$) revealed strong field-induced quasi-static magnetism in the underdoped and Eu doped $(La,Sr)_2CuO_4$ and $La_{1.875}Ba_{0.125}CuO_4$, existing well above T_c and T_N. The susceptibility-counterpart of Cu spin polarization, derived from the muon spin relaxation rate, exhibits a divergent behavior towards $T \sim 25$ K. No field-induced magnetism was detected in overdoped $La_{1.81}Sr_{0.19}CuO_4$, optimally doped Bi2212, and Zn-doped $YBa_2Cu_3O_7$.

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The interplay between magnetism and superconductivity is one of the central subjects in the study of high T_c superconductivity [1]. Recently, remarkable effects have been observed in experiments with high external magnetic fields B_{ext} applied parallel to the c-axis of single crystal specimens. Lake *et al.* [2] found substantial increase of inelastic neutron scattering intensity at low energy transfers and incommensurate wavevectors with increasing field in optimally doped $La_{2-x}Sr_xCuO_4$ (LSCO). More dramatic effects have been observed in underdoped LSCO [3, 4] and in oxygen-doped La_2CuO_{4+y} (LCO) [5], where static spin correlations are enhanced by B_{ext}, as seen by increased elastic intensities of satellite neutron Bragg peaks. NMR experiments in high fields in $Tl_2Ba_2CuO_{6+\delta}$ (Tl2201) [6] and $YBa_2Cu_3O_{7-\delta}$ (YBCO) [7] detected the presence of dynamic antiferromagnetic spin correlations inside vortex cores. There remain, however, many unanswered questions, such as, (a) in which case is the field-induced magnetism static or dynamic ? (b) does it occur exclusively in vortex cores or everywhere in the system ? (c) is this phenomenon generic to all the cuprates or specific to particular series and/or doping ranges ? (d) is the superconducting order parameter affected by the field-induced magnetism ?

Muon spin relaxation (μSR) is a probe well suited to shed some light on these aspects. Following zero-field (ZF) μSR studies on static magnetism of LSCO and YBCO superconductors [8, 9], our recent ZF μSR results [10] elucidated the coexistence of magnetism and superconductivity in $La_2CuO_{4.11}$ (LCO:4.11) and LSCO with $x = 0.12$ (LSCO:0.12), where static magnetism occurs only in a finite volume fraction, forming nano-scale static spin stripe regions with a radius ~ 30 Å (comparable to the in-plane coherence length ξ_{ab}). In μSR measurements with low transverse field (TF) ($B_{ext} = 0.2$ T) in $La_{2-x-y}Eu_ySr_xCuO_4$ (LESCO) [11], we found a tradeoff between the superfluid density and the magnetically ordered volume fraction, which indicates that the superconductivity and static spin ordering occur in microscopically intertwined but spatially separate regions.

TABLE I: Single crystal specimens studied in this work

Composition	Abbreviation	T_c (K)	T_N (K)	V_{Cu} (%)
$La_{1.88}Sr_{0.12}CuO_4$	LSCO:0.12	28	20	40
$La_{1.875}Ba_{0.125}CuO_4$	LBCO	2.5	40	80-100
$La_{1.75}Eu_{0.1}Sr_{0.15}CuO_4$	LESCO	20	22	~50
$La_{1.81}Sr_{0.19}CuO_4$	LSCO:0.19	30	0	0
$YBa_2(Cu_{2.979}Zn_{0.021})O_7$	YBCO(Zn)	80	0	0
$(Bi,Pb)_2Sr_2CaCu_2O_8$	Bi2212	80	0	0

In this paper, we report TF-μSR studies of the effect of high magnetic fields on several cuprate systems, listed in Table 1, using single crystal specimens. The values of their transition temperatures for superconducting state (T_c) and static magnetic order (T_N) in ZF are shown together with the volume fraction V_{Cu} of static Cu moments in ZF determined by ZF-μSR. Refs. [3, 10, 12] describe growth and/or characterization of similar (not identical) crystals with the same nominal compositions made by the groups who prepared the present specimens.

Specimens of dimensions ~ 8 mm x 8 mm x 1 mm are mounted in He gas-flow cryostats with the largest face (ab-plane) perpendicular to the muon beam direction $\hat{z}$, along which the external field was applied with an initial muon spin polarization made perpendicular to $\hat{z}$ using a spin rotator. We used the HI-TIME μSR spectrometer

at TRIUMF to detect high-frequency precession signals. Time resolution and positron trajectories restricted the highest available field to $B_{ext} = 6$ T.

Traditionally, the TF-μSR data in high fields have been analyzed using Fourier Transforms and/or rotating reference frame (RRF) [13, 14]. Neither of them, however, clearly display the existence of multi-component signals. As an alternative method [15], we extracted the amplitude of the signal from very short time intervals (~ 50 ns) by fitting the precession signal to a simple sinusoidal form $A\cos(\omega t+\phi)$. In such a time domain, the amplitude and frequency are practically constant, and the derived amplitude A for each interval represents the muon spin relaxation function in TF, which we call the "envelope" of the precession signal. Figure 1 shows the time evolution of the envelope in several systems.

A significant increase of the relaxation rate with increasing fields is seen in LSCO:0.12 and LESCO at $T \ll T_c$ [Figs. 1(a) and (b); B_{ext} shown in (b)], and also at $T > T_c$ and $T > T_N$ as shown for LSCO:0.12 in Fig. 1(a). We obtained similar results in LBCO. The observed relaxation in these systems at $T < T_N$ is due predominantly to magnetism, since their relaxation rates are much higher than those expected from the magnetic field penetration depth λ. In LSCO:0.12 at $T < T_N$, the envelope shows a clear two-component decay, as demonstrated in the logarithmic plot of Fig. 1(c). In contrast, essentially no field dependence has been observed

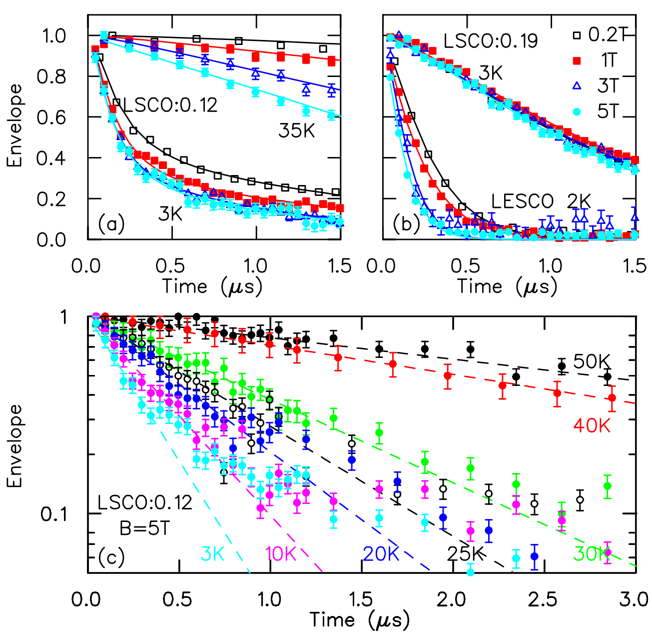

FIG. 1: (color) The envelope of muon spin precession in transverse external fields B_{ext} parallel to the c-axis observed in (a) LSCO:0.12 at $T = 3$ K and 35 K; (b) LSCO:0.19 at $T = 3$ K and LESCO at $T = 2$ K; and (c) LSCO:0.12 in $B_{ext} = 5$ T in a logarithmic plot which demonstrates decay in two-component signals below $T \sim 25$ K.

in LSCO:0.19 (Fig. 1(b)), YBCO(Zn) and Bi2212, in which the relaxation can be explained in terms of the penetration depth λ in the superconducting state. A slight reduction of the relaxation rate at higher fields in YBCO(Zn) and Bi2212 can be attributed to a small increase of B_{ext} relative to H_{c2} [16].

We fit the observed envelope of LBCO, LESCO, and LSCO:0.12 ($T > 25$ K) with a single exponential decay $\exp(-\Lambda t)$ and derived the muon spin relaxation rate Λ, as shown in Fig. 2. In the case of the two-component decay of LSCO:0.12 below T_N, we fit the envelope with a sum of two exponentially decaying functions. The points in Fig. 2(b) for this region (left of the vertical solid line) correspond to the exponential decay rates of the faster component, i.e, the slope of the broken lines in Fig. 1(c), while the points above T = 25 K represent Λ in the single-component fit. These single- and two-component exponential fits in LSCO:0.12 and LESCO are displayed by the solid lines in Figs. 1(a) and (b), which exhibit a good agreement with the observed data.

In addition to large and field-dependent relaxation rate below T_N due to local field from static Cu moments, we see a significant field-dependent relaxation well above T_c

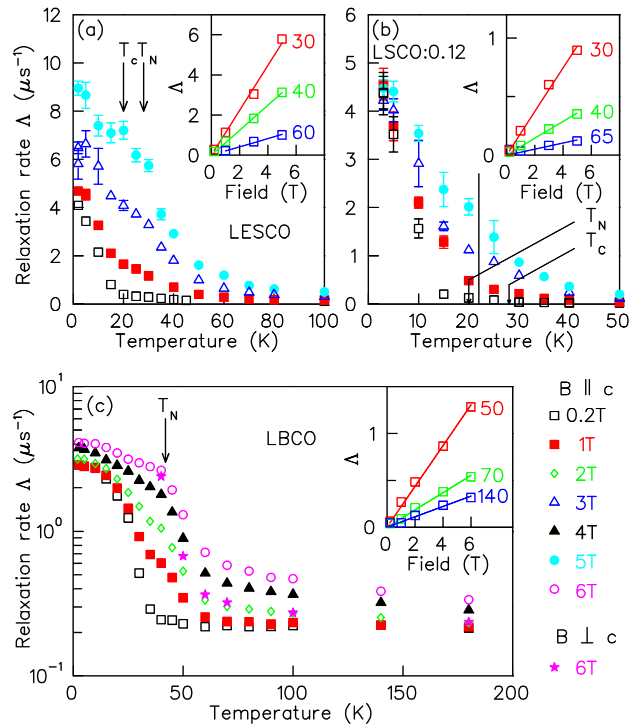

FIG. 2: (color) Muon spin relaxation rate Λ in TF-μSR with B_{ext} applied parallel to the c-axis in (a) LESCO, (b) LSCO:0.12 and (c) LBCO. The purple star represents the results in LBCO with $B_{ext} \perp \hat{c}$. Points of Λ represent the decay rate of a single-component exponential signal, except for $T < 25$ K in LSCO:0.12 where they represent those of the faster component in a two-component exponential signal. The insets show field dependence of Λ at selected temperatures.

and T_N in LESCO, LSCO:0.12 and LBCO. As shown in the inset figures of Fig. 2, the relaxation rates Λ above T_N, after correction for background relaxation due to nuclear dipolar fields and magnet-related inhomogeneous fields, exhibit a linear relation to the external field at a given temperature. In LBCO, we also performed some measurements with crystals cut into a different orientation to have the external field perpendicular to the c-axis, whose results are shown by the purple stars in Fig. 2(c). While Λ exhibits nearly no dependence on the field orientation below T_N, we found a remarkable reduction of the field-induced effect above T_N for $B_{ext} \perp \hat{c}$.

From the slope of the field dependence of Λ for $B \parallel c$-axis, such as those shown in the insets of Fig. 2, we obtained $d\Lambda/dB$, which might be called the "susceptibility-counterpart" since the field-induced relaxation in these system is likely due to static random fields as discussed later. Figure 3(a) shows $dB/d\Lambda$, or the counterpart of inverse susceptibility, in these three systems as a function of temperature. We find that $dB/d\Lambda$ shows nearly linear temperature dependence with the intersect to the horizontal axis at $T \sim 25$ K common to all the three systems. This is a behavior analogous to inverse dc susceptibility of a ferromagnet having the Curie temperature of 25 K.

Using a SQUID magnetometer, we also measured the dc-susceptibility χ_{dc} of our specimens of LSCO:0.12 and LESCO in an external field $B \parallel \hat{c}$ of 0 - 5 T. At $T_c < T < 200$ K, the magnetization M exhibits linear variation with B, with the field-independent slope dM/dB. Figure 3(b) shows the inverse susceptibility $1/\chi_{dc} \equiv dB/dM$ in LSCO:0.12 and LESCO. The large χ_{dc} in LESCO is due

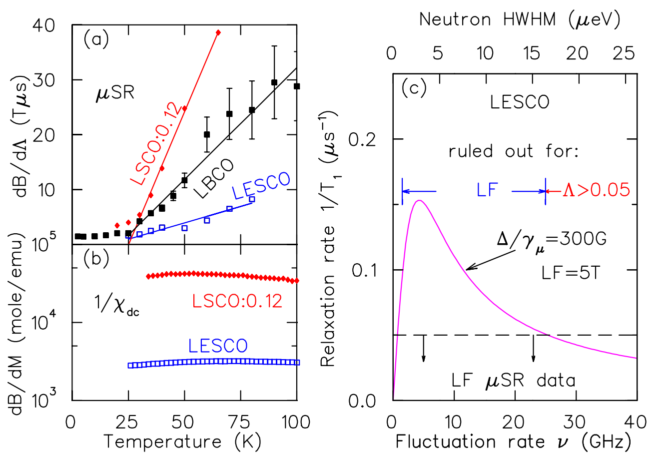

FIG. 3: (color) (a) The inverse "susceptibility-counterpart" $dB/d\Lambda$ derived from the dependence of the relaxation rate Λ in TF-μSR on $B_{ext} \parallel \hat{c} = 0.2$ - 6 T. (b) The inverse of dc-susceptibility $\chi_{dc} \equiv dM/dB$ determined from the magnetization M for $B \parallel \hat{c} = 0.2$ - 5 T. (c) Expected longitudinal relaxation rate $1/T_1$ in LF-μSR for Cu spin fluctuations with the rate ν compared with the LF-μSR results in LESCO at external field of 5 T. ν is also displayed with corresponding energy width (HWHM) in neutron studies.

to the Van Vleck term of Eu^{3+}. In both systems $1/\chi_{dc}$ is nearly independent of temperature, in a remarkable contrast to the "counterpart from μSR" in Fig. 3(a).

In order to study possible dynamic effects, we have also performed μSR measurements of LESCO [and LBCO] under longitudinal field (LF) of $B_{ext} \parallel \hat{c} = 5$ T [4 T] at temperatures between 2 K and 150 K (of 20-30 K intervals) [2.5 and 160 K; 20 K intervals]. We found no relaxation at any temperature, which provides an upper limit of the muon spin relaxation rate $1/T_1 < 0.05\mu s^{-1}$. When Cu moments of ~ 0.5 μ_B/Cu become static, the internal magnetic field of 300 G would be created at the muon site in LESCO and LBCO [10, 11]. For Cu spin fluctuations with the rate ν, we simulated $1/T_1$ in LF of $B_{ext} = 5$ T assuming a Gaussian random internal field with the width $\Delta/\gamma_\mu = 300$ G, where γ_μ denotes the gyromagnetic ratio of the muon spin. The simulation results in Fig. 3(c) exceed the upperlimit of $1/T_1$ observed in LESCO for $\nu =$ 1-25 $\times 10^9$/s, ruling out ν in this region (blue arrows in Fig. 3(c)). When ν is significantly larger than $\nu \sim \gamma_\mu B_{ext}$ which gives the $1/T_1$ maximum, Λ in TF should be nearly equal to $1/T_1$ in LF, contrary to the observed data $\Lambda \gg 1/T_1$. Consequently, the fast fluctuation $\nu > 25 \times 10^9$ /s cannot explain observed results. Combined results in LF and TF now indicate that $\nu > 10^9$ /s is incompatible with our data.

Thus, we consider that the observed relaxation above T_N is due to quasi-static random fields induced by the external field. Since this effect does not appear in the uniform susceptibility, it should be related to Cu spin polarization with finite wavevector component q, presumably originating from (possibly short ranged) stripe spin correlations which are stabilized below T_N. The dependence on the crystal orientation in LBCO rules out "polarization of dilute magnetic impurities" as a possible explanation. Good fit of the envelope to single exponential decay above T_N suggests that the observed phenomenon is not confined to a small volume fraction of the system.

Regarding the two component signals in LSCO:0.12 below T_N, the signal with fast (slow) relaxation must be due to volume fractions with (without) static magnetic order. In Fig. 4(a), we show the amplitude fraction V_μ of muons involved in the fast relaxation. The volume fraction V_{Cu} of static Cu moments is slightly smaller than V_μ, since muons stopped near the edge, but outside, of the "magnetic islands" are also depolarized. After converting V_μ into V_{Cu} using Fig. 8 of ref. [10], we obtained $(1 - V_{Cu}) \equiv V_{sc}$. (Different models for the shape of magnetic regions would yield qualitatively similar behavior of V_{Cu}.) If one assumes that the slow relaxation is due to the superconducting region, V_{sc} represents the superconducting volume fraction. Figure 4 shows that both $V_{sc}(T \rightarrow 0)$ and T_c decreases with increasing field, but their ratio is nearly independent of the field. Similar proportionality between T_c and the superconducting volume fraction [17] has been observed in the case of varying T_c

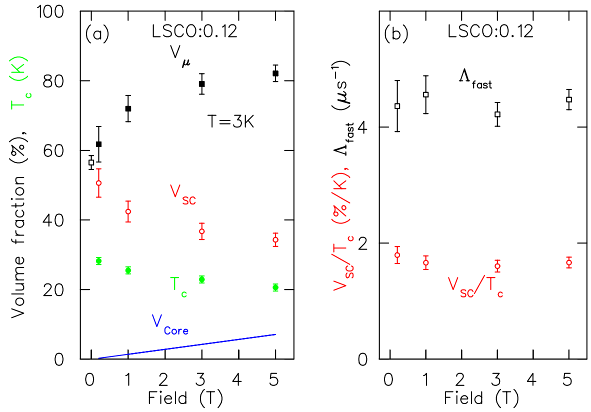

FIG. 4: (color) (a) The volume fractions of the fast muon decay signal V_μ and superconducting region V_{sc} and T_c in LSCO:0.12, and the core region V_{core} for a superconductor with ξ_{ab} = 23 Å. (b) The decay rate Λ_{fast} at T = 3 K and V_{sc}/T_c in LSCO:0.12.

with (Cu,Zn) substitution in LSCO systems [18] and increasing Eu concentration in LESCO [11].

In LSCO:0.12, the relaxation rate Λ of the fast-decay component is nearly independent of the field at $T \leq 5$ K, as shown in Fig. 2(b) and 4(b). This implies that the external field increases the magnetic volume fraction but does not change the moment size. The blue solid line V_{core} in Fig. 4(a) represents volume fraction of the vortex core region for superconductors having the in-plane coherence length $\xi = 23$Å. We can rule out the "vortex cores with full static magnetism" at T = 2-3 K in YBCO:Zn, Bi2212 and LSCO:0.19, in which the observed envelopes in B_{ext} = 5-6 T indicate that muon spins involved in such "fast decay" is less than 1% in volume. Static magnetism with much smaller field ($\sim$ 15 G) in the vortex core, proposed for an underdoped YBCO [13], still remains as a possibility in these three systems.

The strong field dependent effects above and below T_N were observed only in the systems which exhibit static magnetism in ZF in partial or neally full volume fraction V_{Cu} (see Table 1.). Thus, the field-induced relaxation is not generic to all the cuprate systems but is confined to systems having competing magnetic state very close in free energy to their superconducting state. The stronger effect for the field applied perpendicular to the CuO_2 planes in LBCO suggests that this phenomenon could be related to suppression of superconductivity caused by the field. Though the superconducting T_c's are rather low in underdoped LSCO:0.12, LBCO and LESCO systems, the "dynamic superconductivity"[17, 19] detected by the Nernst effect [20] might extend up to $T \sim 150$ K ("Nernst region"). The suppression of dynamic superconductivity could favor competing antiferromagnetism [21].

Recently, a very small diamagnetic magnetization M_{dia} was detected in Bi2212 in the Nernst region above T_c [22]. The field and temperature dependences of M_{dia} follow behavior similar to that of Λ in μSR. This suggests a possibility that the observed relaxation above T_c is due to the dynamic supercurrent screening B_{ext}. However, if we assume $\Lambda = \alpha M_{dia}$ and obtain the proportionality factor α using the results in Bi2212 near T = 0, that factor gives the value of Λ more than 10 times smaller than the observed value for a reasonable estimate of M_{dia} above T_c in LSCO and LESCO using the Nernst results. Furthermore, it is difficult [19] to expect the dynamic diamagnetism to have the time scale slower than $t \sim 1\mu$s, required for a flux vortex lattice to produce field inhomogeneities observable in μSR. These difficulties have to be resolved before the dynamic screening scenario is adopted.

In conclusion, our μSR measurements in high TF revealed field-induced magnetism in LSCO:0.12, LESCO, and LBCO existing in a wide range of temperature above and below T_c and T_N. This phenomenon is not common to all the cuprate superconductors, but rather confined to those systems having magnetically ordered state closely competing with the superconducting state.

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* author to whom correspondences should be addressed

[1] See, for example, papers in the Proceedings of the 2002 Williamsburg HTSC Workshop, ed. by A.J. Millis, S. Uchida, Y.J. Uemura, Solid State Commun. **126** (2003).
[2] B. Lake *et al.*, Science **291**, 1759 (2001).
[3] S. Katano *et al.*, Phys. Rev. B **62**, R14677 (2000).
[4] B. Lake *et al.*, Nature **415**, 299 (2002).
[5] B. Khaykovich *et al.*, Phys. Rev. B **66**, 014528 (2002).
[6] K. Kakuyanagi *et al.*, Phys. Rev. Lett. **90**, 197003 (2003).
[7] V. F. Mitrovic *et al.*, Nature **413**, 501 (2001).
[8] A. Weidinger *et al.*, Phys. Rev. Lett. **62**, 102 (1989).
[9] Ch. Niedermayer et al., Phys. Rev. Lett. **80**, 3843 (1998).
[10] A. T. Savici *et al.*, Phys. Rev. B **66**, 014524 (2002).
[11] K. M. Kojima *et al.*, Physica B **326**, 316 (2003).
[12] J. M. Tranquada *et al.*, Nature **429**, 534 (2004)
[13] R. I. Miller *et al.*, Phys. Rev. Lett. **88**, 137002 (2002).
[14] R. Kadono *et al.*, Phys. Rev. **B69**, 104523 (2004).
[15] Y. J. Uemura *et al.*, Solid State Commun. **31**, 731 (1979).
[16] J. E. Sonier *et al.*, Phys. Rev. Lett. **83**, 4156 (1999).
[17] Y.J. Uemura, J. Phys. Condens. Matter **16**, S4515 (2004).
[18] B. Nachumi *et al.*, Phys. Rev. Lett. **77** (1996) 5421.
[19] P. W. Anderson, cond-mat/0504453.
[20] Y. Wang *et al.*, Phys. Rev. **B64**, 224519 (2001).
[21] S. Sachdev and E. Demler, Phys. Rev. **B69**, 144504 (2004).
[22] Y. Wang *et al.*, cond-mat/0503190.